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A continuous time random walk approach to transient flow in heterogeneous porous media

Andrea Cortis[1] and Christen Knudby[2]

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[1] We propose a new physical interpretation of the diffusion process for the piezometric head $h(x, t)$ in heterogeneous aquifers based on the continuous time random walk (CTRW) theory. For the typical heterogeneities considered in this work, we find that a CTRW based diffusion equation for $h(x, t)$ provides better fits to the transient flow simulations than the classical diffusion equation (DE). The DE is found to be a special case of the CTRW diffusion equation. The results of this work have clear implications for the interpretation of pumping tests and what information can be extracted from them.

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[2] Well testing is an essential tool for groundwater aquifer assessment, the key idea being that hydraulic properties can be derived by means of transient drawdown and/or flux measurements. The commonly accepted mathematical model for groundwater flow in porous formations is based on the well known Darcy law, which relates the fluid flow to the gradient of the piezometric head, $h(x, t)$ $[L]$. Together with a mass balance and the assumptions of small compressibility, constant porosity and permeability, and small head gradients, it is possible to derive a partial differential equation of parabolic type (diffusion equation) for h

$$\partial_t h(\mathbf{x}, t) = \nabla(\alpha(\mathbf{x})\nabla h(\mathbf{x}, t)), \tag{1}$$

where $\alpha = K/S$ is the hydraulic diffusivity $[L^2/T]$, S is the specific storage $[1/L]$, and K is the hydraulic conductivity $[L/T]$. We note that the characteristic time τ for the diffusion of h is determined solely by the value of α. Simple analytical solutions for (1) exist for different boundary conditions (BCs), such as leaky aquifers, well skin effects, and wellbore storage. Almost all these solutions are based on the simplifying assumption of homogeneity, i.e, α is treated as a constant characteristic of the particular geological formation. Current hydrogeological practice relies heavily on the Jacob, Theis and Hantush methods which are all derived from the analytical solutions of (1). Comprehensive reviews of these methods can be found in any standard textbook on the interpretation of pumping tests [e.g., *Streltsova*, 1988].

[3] Often, however, pumping tests show significant deviations from these classical textbook solutions [*Raghavan*, 2004]. Some of these deviations can be attributed to an imperfect knowledge of the BCs for the flow, while others are related to the unknown heterogeneity of the aquifer.

[4] In this work we shall focus exclusively on the effects of the heterogeneities. Heterogeneity-related deviations from the classical solutions of (1) can be more or less pronounced depending on the conductivity contrast between adjacent regions and on their degree of connectedness [e.g., *Knudby and Carrera*, 2005]. This leads to the problem of defining appropriate scale-up procedures to derive macroscopic flow and transport coefficients from the knowledge of the underlying heterogeneity. Early works by *Dagan* [1982], *Naff* [1991], and *Indelman* [1996] recognized and analyzed the importance of heterogeneities for the transient flow problem. In the following, we will refer to as "anomalous" the deviations of the effective drawdowns/fluxes with respect to the classical solution for a homogeneous field (Gaussian regime).

[5] An effective way of accounting for the effect of heterogeneity in transport problems is the continuous time random walk (CTRW) method. In hydrogeological applications, CTRW has proved to be highly successful for the treatment of the impact of heterogeneities on transport of contaminants in porous and fractured systems, both at the laboratory and field scale [*Berkowitz and Scher*, 1995, 1997; *Cortis et al.*, 2004a]. The objective of this study is to extend the physical picture of the CTRW theory to investigate the problem of transient flow in heterogeneous aquifers.

[6] Our point of departure is the master equation (ME) (see, e.g., *Oppenheim et al.* [1997, chapter 3] and *Berkowitz et al.* [2006] for a more hydrogeologically oriented exposition). The ME is a phenomenological first-order differential equation describing the time evolution (Markov process) of the probability of a system to occupy each of a (discrete or continuous) set of states. The discrete form of the ME for $h(x, t)$ reads

$$\partial_t h_i(t) = w_{ij} h_j(t), \tag{2}$$

where $h_i(t) \equiv h(\mathbf{x}_i, t)$ and w_{ij} is the transition rate $[1/T]$ of h from point $\mathbf{x}_j$ to $\mathbf{x}_i$. Einstein's summation convention on

[1]Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California, USA.
[2]Schlumberger-Doll Research, Cambridge, Massachusetts, USA.

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repeated indices is assumed. Equation (2) has been utilized widely in the physics and chemistry literature [e.g., *Klafter and Silbey*, 1980]. The ME (2) represents here the balance equation for the fluid total energy density. Note that (2) does not assume a ''Darcy-like'' flux behavior for the evolution of $h(\mathbf{x}, t)$.

[7] To better appreciate the physical meaning of the transition rates w_{ij}, we derive their precise expression for the simple case of diffusion of h in a completely homogeneous medium. Consider the discretized form of (1)

$$\partial_t h_i(t) = \left(\mathsf{D}_{ik}\alpha_k \mathsf{D}_{kj}\right) h_j(t), \tag{3}$$

where the spatial gradient has been approximated by the linear operator (matrix) $\partial_x \simeq \mathsf{D}_{ij}$. Identification of the right-hand side of (3) and (2) yields the expression for the transition rates

$$w_{ij}^{(x)} = \mathsf{D}_{ik}\alpha_k \mathsf{D}_{kj}. \tag{4}$$

Equation (4) states that the transition rates of the ME for a completely homogeneous system are determined by a spatial component depending on the precise position of the sites and, importantly, by a single characteristic time, τ, given by the diffusivity coefficient $\alpha \sim L_s^2/\tau$, where $L_s \ll L_x$. L_x is the characteristic length scale at which the variation of the diffusivity is supposed to be known. Having shown that the ME (2) is just another way of expressing the transient evolution of the piezometric head, h, we can assume (2) as the starting point for our generalization.

[8] For heterogeneous media, the range of characteristic times is wider and depends on the spatial distribution of $\alpha(x)$. The determination of $w_{ij}^{(x)}$ requires detailed knowledge of the system at length scales L_x, i.e., the characterization of the heterogeneities on all length scales that influence the calculation of the flow field. Below this characteristic length scale we must resort to a statistical description of $\alpha(s)$ and hence to a probability distribution of $w_{ij}^{(s)}$ at $L_s \ll L_x$.

[9] To implement this probabilistic approach we consider the ensemble average of (2) [*Klafter and Silbey*, 1980]. We first rewrite (2) in Laplace space

$$u\tilde{h}_i(u) - h_i^0 = w_{ij}^{(s)}\tilde{h}_j(u), \tag{5}$$

where the tilde indicates the Laplace transform $\tilde{h}_i(u) \equiv \mathcal{L}[h_i(t)] \equiv \int_0^\infty h_i(t)\exp(-ut)dt$, and u is the Laplace variable. The ensemble average, [[]], of the ME can be written as [*Klafter and Silbey*, 1980]

$$u\left[\left[\tilde{h}_i(u)\right]\right] - h^0 = \tilde{W}_{ij}(u)\left[\left[\tilde{h}_j(u)\right]\right], \tag{6}$$

and is referred to as the ''generalized master equation'' (GME). The ensemble average of the small-scale transition rates not only depends on space, but also on time (i.e., the generalized transition rates $\tilde{W}_{ij}$ depend on u). We will assume that the spatial and temporal components of $\tilde{W}_{ij}(u)$ can be decoupled in the form

$$\tilde{W}_{ij}(u) \equiv \tilde{M}(u) w_{ij}^{(x)}, \tag{7}$$

i.e., we assume that all the information on the unknown characteristic times for the diffusion at the scale L_s is lumped into a memory function $M(u)$, whereas the spatial influence of the heterogeneities is mapped onto time-independent transition rates $w^{(x)}$ at the scale L_x. The ensemble average operator introduces a memory effect in the structure of the PDE and thus explicit flow calculations on multiple realizations of the diffusivity field are not required.

[10] Inserting (7) and (4) into (6) and going to the continuum limit we obtain

$$S\left(u\left[\left[\tilde{h}_i(u)\right]\right] - h_i^0\right) = -\partial_x \tilde{q}(x, u), \tag{8}$$

where the generalized flux $\tilde{q}(x, u)$ is written as

$$\tilde{q}(x, u) = -\tilde{M}(u) K(x) \partial_x\left[\left[\tilde{h}_j(u)\right]\right]. \tag{9}$$

Equation (8) represents the evolution of the mean piezometric head; the evolution of the variance of $[[\tilde{h}_i(u)]]$ is a subject of current investigation. The measured piezometric head is thus the result of an average over all the possible (unknown) small-scale heterogeneities. In what follows, we will drop the cumbersome ensemble average [[]] notation for the piezometric head. Equation (8) contains a large-scale variation for $K(x)$ and in this respect is similar to the hybrid model for transport proposed by *Cortis et al.* [2004a].

[11] *Klafter and Silbey* [1980] proved that the GME is equivalent to a CTRW equation. We can therefore exploit the results valid for CTRW and write the memory function $\tilde{M}(u)$ as [*Dentz et al.*, 2004]

$$\tilde{M}(u) \equiv \bar{t}u \frac{\tilde{\psi}(u)}{1 - \tilde{\psi}(u)}, \tag{10}$$

where $\psi(t)$ is the classical CTRW transition time distribution for the transfer of h, and is a characteristic time. The physical interpretation of $\psi(t)$ in the context of flow is straightforward: it represents the probability rate that the total fluid energy density (the piezometric head, h) is transferred to a neighboring location. In other words, one should not think of actual ''jumps'' of discrete quantities as for instance in the case of the electron hopping transport problem, but rather in terms of rates of continuous quantities. In this respect, the discrete forms of the ME and GME are only convenient tools to derive the continuous partial differential equation (8). The explicit relationship between $w_{ij}^{(s)}$ and $\psi(t)$ is given by *Berkowitz et al.* [2006, Appendix B].

[12] Various forms of $\psi(t)$ have been proposed in the literature to conveniently describe the effect on transport of heterogeneities typically encountered in geological materials [*Cortis et al.*, 2004b]. In this work, we will focus on three expressions for $\psi(t)$, namely, the decaying exponential

$$\psi(t) = \exp(-t), \tag{11}$$

the truncated power law (TPL)

$$\psi(t) = \frac{1}{\left(t_1\, \tau_2^{-\beta}\, \exp\left(\tau_2^{-1}\right) \Gamma\left(-\beta, \tau_2^{-1}\right)\right)} \frac{\exp(-t/t_2)}{(1 + t/t_1)^{1+\beta}}, \tag{12}$$

and the modified exponential (ETA)

$$\psi(t) = \eta_3 F_3 \left[{}^{1,1,1}_{2,2,2} ; -\tau \right] e^{-\eta \tau_4 F_4 \left[{}^{1,1,1,1}_{2,2,2,2} ; -\tau \right]}. \tag{13}$$

A thorough discussion on these three probability transition rates are given by *Berkowitz et al.* [2006]. It will be sufficient here to recall that $\mathcal{L}[\exp(-t)] = 1/(1 + u)$, and thus $\tilde{M}(u) = 1$, i.e., the classical DE (1) reduces to a special case of the CTRW equation (8). The other two forms for $\psi(t)$ lead to ''anomalous'' diffusive transport. The generalized flux, $\tilde{q}(x, u)$, reduces to the classical Darcy law for homogeneous media (i.e., when $\tilde{M}(u) = 1$), or for asymptotic steady state conditions, i.e., when $\tilde{M}(u) \rightarrow 1$ as is the case for the expressions in (12) and (13). The solution of (8) is obtained analogously to what described by *Cortis and Berkowitz* [2005].

[13] The space-time nonlocal nature of (unconditional) mean transient flow has been previously recognized by others, most notably in the works of *Hu and Cushman* [1994] and *Indelman* [1996]. These authors used perturbation analysis to formulate an effective Darcy law and derive a mean flux term that is nonlocal, forming a convolution integral in space-time of a kernel with the mean head gradient [*Tartakovsky and Neumann*, 1998; *Ye et al.*, 2004]. Our analysis is fundamentally different from the aforementioned works.

[14] Our numerical results clearly indicate that the effect of the heterogeneities considered here can be lumped into a nonlocal-in-time memory term; the localization in space of the ensemble averaged transition rates is thus fully justified.

[15] Consider the question, Given a heterogeneous spatial distribution of $\alpha(s)$, is it possible to define an ''equivalent'' macroscopic diffusivity $\bar{\alpha}$ at the scale L_x for the whole domain? To answer this question, we adopt a computational point of view, i.e., we assume that we know a priori the structure of the heterogeneity field. We then simulate a transient flow situation by means of a standard flow simulator. Finally we fit the time evolution of some average quantity by means of (8) with constant $\bar{\alpha}$. Here we consider two-dimensional parameter distributions to illustrate in a simple way the effect of heterogeneity on the diffusion of hydraulic head.

[16] Drawdown measurements can, in theory, always be translated into flux. We therefore present our results in terms of integrated flux without loss of generality. The main results of our development do not depend on the particular choice of BCs and more realistic 3D scenarios can be implemented in a straightforward manner to also include pumping or injection wells as sources or sinks in (8).

[17] Two typical examples of the 2-D conductivity distributions used in this study are shown in Figure 1. The distributions of $Y = \ln(K)$ are multi-Gaussian with a variance of $\sigma_Y^2 = 15.9$, and a geometric mean $\langle Y \rangle^{\mathrm{G}} = 0$. The employed variograms are Gaussian with an isotropic range of 64 grid cells. The domains cover 128×128 grid cells. In Figure 1 the axes lengths have been normalized, $L_x = 1$. The distributions were generated using SGSIM [*Deutsch and Journel*, 1997]. S is assumed constant and equal to 1. The distributions of diffusivity and conductivity are therefore

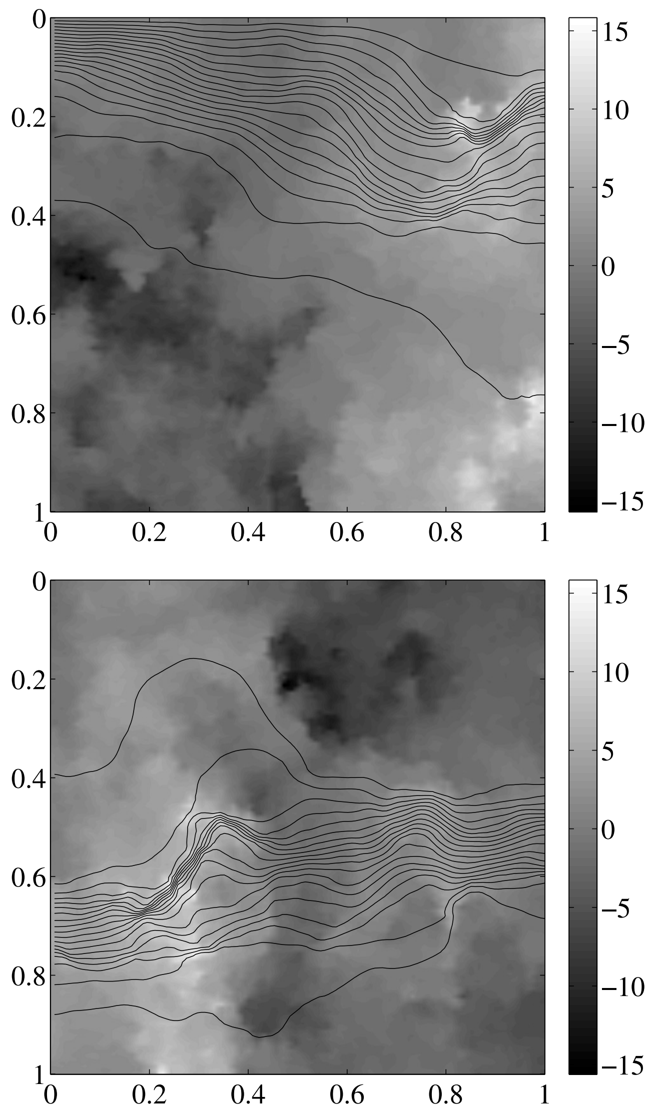

Figure 1. Typical realizations of a multi-Gaussian field. The scales on the right represent $Y = \ln(K)$. The solid lines represent the streamlines for the flow problem.

identical. The arithmetic, geometric, and harmonic mean are the same for both domains and equal to $\langle \alpha \rangle^{\mathrm{A}} = 1782$, $\langle \alpha \rangle^{\mathrm{G}} = 1$ and $\langle \alpha \rangle^{\mathrm{H}} = 0.56 \times 10^{-3}$, respectively.

[18] The average flow is assumed to be horizontal in the (x_1, x_2) plane. The initial condition is $h(x_1, x_2, 0) = 1$. We impose a no-flow BC on the top, $\partial_{x_2} h(x_1, 0, t) = 0$ and bottom, $\partial_{x_2} h(x_1, 1, t) = 0$, boundaries. A Dirichlet BC is imposed on the left, $h(0, x_2, t) = 1$, and right $h(1, x_2, t) = 0$. The transient flow corresponding to these BCs was simulated by means of MODFLOW-2000, version 1.6 [*Harbaugh et al.*, 2000]. The streamlines for domains A and B are reported in Figure 1. The system relaxes toward a steady state situation corresponding to an imposed macroscopic unit piezometric head gradient. This yields a time-dependent Darcy flow macroscopically directed from left to right. The integral of the flow on the left boundary is defined as $Q(t) = -\int_0^1 K(0, x_2)\, \partial_{x_1} h(0, x_2, t)\, dx_2$. In Figure 2 we report the normalized flux $\hat{q}(t) = Q(t)/Q(\infty)$.

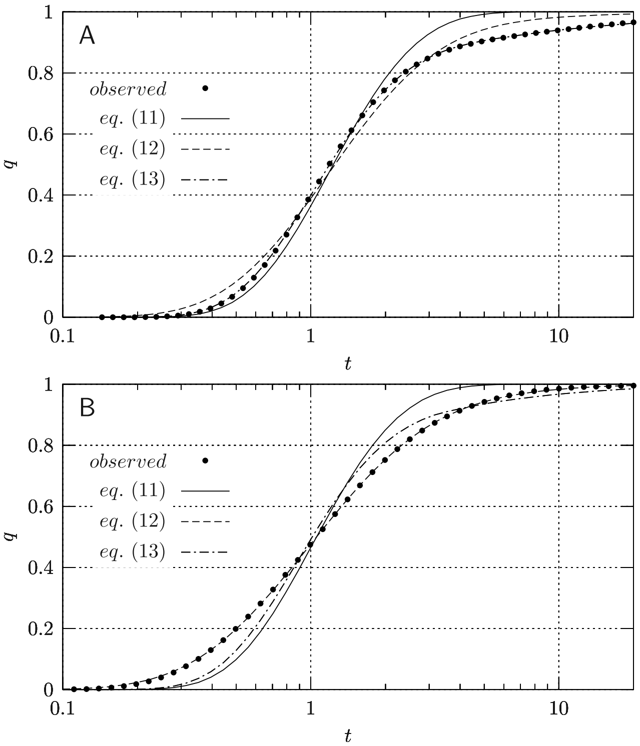

Figure 2. Normalized flow q as a function of time t due to a sudden drop in the piezometric head on the right boundary. The dots ("observed") represent the numerical solution of (1), $\hat{q}(t)$, for the heterogeneous domains in Figure 1. The solid, dashed, and dash-dotted lines represent the best fit of $\hat{q}(t)$ with the classical diffusion equation, (i.e., (8) with (11), (8) with (12), and (8) with (13), respectively).

[19] The solution of the 1D form of (8) for the initial condition $h(0, t) = 1$ and BCs $h(0, t) = 1$ and $h(1, t) = 0$ gives an expression for the Laplace transform of the normalized flux, $\tilde{q}(x, u)$ equal to [e.g., *Carslaw and Jaeger*, 1986]

$$\tilde{q}(x,u) = S^{-1}\frac{\xi}{u}\frac{\cosh(\xi x)}{\sinh(\xi)}, \quad \text{where} \quad \xi = \sqrt{\frac{u}{\alpha\tilde{M}(u)}}. \tag{14}$$

Equation (14) is evaluated at $x = 1$ and then numerically inverted back to the time domain to get the evolution of $q(t)$.

[20] We calculated the best fit parameters of (14) for the three models of $\psi(t)$ in (11), (12), and (13) that minimize the norm $||q(t) - \hat{q}(t)||$. Deviations from (1) (i.e., from (8) with (11) are observed at both early and late arrival times. This means that the heterogeneities influence how fast the piezometric head at one location affects its neighboring locations. For both domains, the best value for $\bar{\alpha} \approx 0.12$. For Domain A, the ETA function in (13) gives the best results with $\eta = 6.28 \times 10^{-2}$ and $\bar{\alpha} = 1.99$, whereas the TPL function gives a less good fit. The situation is reversed for Domain B, where the TPL function gives a better fit than the ETA function. The best fit parameters are in this case $\bar{\alpha} = 1.24$, $\beta = 0.94$, $\tau_2 = 31.68$, and $\tau_1 \sim 0$. Note that the magnitude of the truncation time τ_2 compares well with the total breakthrough time $t_{\max} \sim 20$, which indicates that the system is converging toward an asymptotic Gaussian regime. The fitted values of $\bar{\alpha}$ are much larger than the value of $\bar{\alpha}$ in (1), and comparable with the geometric mean $\langle\alpha\rangle^{\mathrm{G}} = 1$. This is due to the heterogeneity which allows the transient signal to cross the field faster than for the corresponding homogeneous field [e.g., *Knudby and Carrera*, 2006].

[21] We have tested 50 realizations with the same geostatistical parameters adopted for domains A and B. With a few exceptions, we were able to fit all the realizations with (12) or (13). The difference in fitting function between the two realizations may be attributed to a different degree of channeling for the different realizations. In fact, streamlines for Domain A seem to be slightly more regularly spaced than for Domain B, where a more distinct power law behavior is observed. We are currently testing these hypotheses more in depth using a larger set of realizations. Equation (13) was derived by *Cortis et al.* [2004a] under the assumption of exponential decay of the spatial autocorrelation for the Stokes velocity field. This suggests that (9) can be upscaled from the Stokes equations, analogously to what is done with the frequency-dependent dynamic permeability [e.g., *Lévy and Sanchez-Palencia*, 1977]. The fact that (13) is found so commonly to fit our transient flow realizations might derive from our assumption of exponential decay for the spatial autocorrelation of $\ln(K)$. The relationship between the various fitting parameters for (12) or (13) and the geostatistical parameters used to generate the various realizations and the resulting parameter distribution characteristics, in particular the connectivity, is also under investigation.

[22] To conclude this communication, we briefly analyze the relationship between our findings and the problem of the transport of contaminants in geological formations. Introducing the expression for the Darcy's velocity in (9) into the classical advection equation for the (Laplace transformed) bulk concentration $\tilde{c}(x, u)$ we obtain

$$u\tilde{c}(x,u) - c_0(x) = -\tilde{M}(u)K(x)\partial_x\tilde{h}(x,u)\partial_x\tilde{c}(x,u) \tag{15}$$

which tells us that anomalous transport of tracers in heterogeneous media can happen also without recourse to the local dispersion effect. Clearly, the latter cannot be neglected in any realistic situation. The complete transport equation therefore needs to be written as

$$\begin{aligned} u\tilde{c}(x,u) - c_0(x) = &- \tilde{M}(u)\tilde{M}'(u)K(x)\partial_x\tilde{h}(x,u) \\ &\cdot \partial_x(\tilde{c}(x,u) - d\partial_x\tilde{c}(x,u)) \end{aligned} \tag{16}$$

where d is the local dispersivity $[L]$ and $\tilde{M}'(u)$ is the memory function for the transport problem [*Berkowitz et al.*, 2006]. Equation (16) allows the calculation of the nonlocal-in-time effect of the heterogeneities of the contaminant transport in a time-dependent velocity field. The relationship between the $\tilde{M}(u)$ and $\tilde{M}'(u)$ parameters is an open question, which can be resolved only experimentally.

[23] Note that the diffusion problem treated here is mathematically equivalent to other problems found in other geophysical applications like heat transfer and electromagnetism. The coupling of these geophysical problems with the flow and transport processes is a subject of current research.

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A. Cortis, Earth Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, CA 94720, USA. (acortis@lbl.gov)

C. Knudby, Schlumberger-Doll Research, 320 Bent Street, Cambridge, MA 02141, USA.